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(54) A HEAT-AND PRESSURE-SENSITIVE ADHESIVE SHEET AND METHOD OF PRODUCING THE SAME

(71) We, UNITIKA LIMITED, of 50, Higashi Hommachi, 1-chome, Amagasaki-shi, Hyogo-ken, Japan and UNITIKA SPARKLITE CO. LTD., of 22, Hatchomae Ogawa, Chiyokawa-cho, Kameoka-shi, Kyoto-fu, Japan, both Japanese companies do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to an air-impermeable heat-and pressure-sensitive adhesive sheet constructed so that it will offer superior effects when bonded to an object.

Conventionally, when a heat and pressure-sensitive adhesive sheet (hereinafter abbreviated to sheet) having a heat-and pressure-sensitive adhesive layer used for a mark, signboard, retro-reflecting traffic sign or the like, is bonded to a backing object, a vacuum applicator is generally used. Thus, with the sheet closely contacted with the object, evacuation by the applicator causes a diaphragm to press the sheet against the object and while the resulting air bubbles between the surfaces of the object and adhesive layer are being removed, the sheet is bonded to the object by the application of heat and pressure. In this case, some of the air bubbles are left behind between the bonded surfaces while a very small amount of solvent contained in the adhesive is gasified to form small bubbles which then remain between the bonded surfaces.

To provide improvements in this respect, an arrangement has been suggested in which a number of tiny air passage holes are provided which extend through all layers of the sheet so that the air bubbles and solvent vapor bubbles can be removed therethrough during the application of heat and pressure. With these known arrangements, however, the provision of tiny air passage holes results in a new problem. That is, since such tiny air passage holes are formed by punching the sheet so that they extend through the sheet from the surface to the back thereof, the peripheries of the tiny air passage holes on the back of the sheet present convexed surfaces while their peripheries on the surface of the sheet present concaved surfaces having a diameter 3—5 times as large as that of the holes. Therefore, the more densely the tiny air passage holes are punched, the higher is the tendency of said concaved surfaces to trap ink during printing operations, blocking up the tiny air passage holes to impair their merits and deteriorate the quality of the printing. In particular, when the sheet is a retro-reflecting sheet, the sheet has a glass bead layer or other reflecting layer as its surface layer or intermediate layer, so that the punching of tiny air passage holes extending through all layers results in the retro-reflectivity being greatly decreased and the printed surface being deteriorated by many dense tiny air passage holes and released beads. Further, if the sheet formed with tiny air passage holes is bonded to a sharply curved object, it is greatly elongated along the curved surface, with the presence of the tiny air passage holes causing the sheet to crack starting at the holes, thus detracting from the commercial value.

The present invention has been accomplished as a result of our intensive researches to improve the mechanism for degassing from between the bonded surfaces of the conventional sheet as described above.

According to the present invention there is provided an air impermeable heat-and pressure-sensitive adhesive sheet provided on its back surface with a heat-and pressure-sensitive adhesive layer, wherein the surface of said adhesive layer is provided with one or more elongate ridges or recesses and wherein at least one end of such ridge or recess intersects the end edge of the sheet.

The present invention will now be

described with reference to the accompanying drawing, in which:

Figures 1 and 2 are sections of different embodiments of heat-and pressure-sensitive sheets according to the invention;

Figures 3—1, 3—2, 4—1 and 4—2 are sections of embodiments, showing sheets before and after being bonded; and

Figures 5—1—3 show the surfaces of different embodiments.

A sheet according to the invention comprises an air-impermeable plastics sheet 1 and a heat-and pressure-sensitive adhesive layer 2 which is applied to the back of said sheet 1 and which has almost no stickiness and has a suitable hardness at ordinary temperature but which will melt at 60°C—150°C. The surface of the adhesive layer 2 is provided with a series of recesses of any suitable cross-section such as an arc *3a*, triangle *3b* or rectangle *3c*, as shown in Figure 1, or ridges 4 of any suitable cross-section such as an arc *4a*, triangle *4b*, or rectangle *4c*, as shown in Figure 2, spaced at predetermined intervals. At least one of the respective ends of such recesses or ridges intersects the lateral edge of the sheet 1.

When a vacuum applicator is used for bonding the heat-and pressure-sensitive adhesive sheet to an object, within said applicator (not shown), the sheet 1 is placed on an object 5 with its adhesive layer 2 contacted with the sheet receiving surface 6 of the object, whereupon the applicator is evacuated with the diaphragm thereof covering said sheet 1. As a result, the diaphragm presses the sheet 1 against the object 5. When the interior of the applicator is heated to a temperature above the melting point of the adhesive layer 2 in such condition, the adhesive layer 2, which is pressed between the sheet 1 and the object, melts. After the lapse of a suitable period of time, the heating is stopped to allow the adhesive layer 2 to set fused to the sheet receiving surface 6, whereby the sheet 1 is strongly bonded to the object 5 (as for the time when the evacuated space is allowed to resume its atmospheric pressure condition, it may be simultaneous with or a suitable period of time after the stoppage of the heating). In the sheet 1 according to the present invention, while bonding it to the object 5 by the conventional heat and pressure method as described above the following merits will be obtained.

Thus, Figure 3—1 shows the sheet 1 of the invention provided with recesses *3a* of arcuate cross-section. When it is brought into abutment against the object 5 from the Figure 3—1 to the Figure 3—2 condition, the recesses *3a* define passageways 7 between the sheet receiving surface 6 and the adhesive layer 2. Further, Figure 4—1 shows the sheet of the invention provided with ridges *4a* of arcuate cross-section. When it is brought into pressure contact with the object 5 from the Figure 4—1 to the Figure 4—2 condition, adjacent ridges *4a* cooperate with an intervening adhesive layer bulging deformed portion *2d* to define passageways *7a* and *7b* between the sheet receiving surface 6 and the adhesive layer 2. In addition, if the distance between adjacent ridges is short, only a single passageway will be defined therebetween, since no deformed portion *2d* abutting against the sheet receiving surface 6 is formed. Each of the passageways 7, *7a* and *7b* is continuous, with its ends opening to the outside at the end edges of the sheet or to the evacuated chamber of the applicator if the sheet is set therewithin. Thus, when the heating and pressing operation is performed in the manner described above, any air bubbles produced between the abutting surfaces of the sheet receiving surface 6 of the object and the adhesive layer 2 and the solvent gas bubbles produced from within the adhesive layer owing to the heating will pass through the passageways 7, *7a* and *7b* until they are removed through the ends of the passageways from between the sheet 1 and the object 5. Until about the time when the adhesive layer 1 reaches its melting point and begins to melt as the heating continues, the degassing action is maintained in order to retain the passageways 7, *7a* and *7b*. As the melting of the adhesive layer 2 continues, the passageways 7, *7a* and *7b* slowly disappear as a result of the degassing action and pressing action due to the diaphragm sheet without leaving air bubbles behind, and the adhesive layer 2 becomes a molten layer of substantially uniform thickness filling the clearance between the sheet 1 and the sheet receiving surface 6 of the object. The surface of the sheet 1 after being bonded under heat and pressure, therefore, is smooth, with possible unevenness due to the previous presence of the passageways 7, *7a* and *7b* not manifesting itself.

The tiny recesses or ridges to be provided on the surface of the adhesive layer of the sheet according to the present invention are in the form of parallel straight lines as shown in Figure 5—1, parallel curves such as parallel waves as shown in Figure 5—2 or a grid network as shown in Figure 5—3. Such recesses and ridges may be provided in combination, and they are parallelly spaced at desired intervals. If one or both ends of each recess or ridge intersect the lines along which the sheet is cut when used, the degassing effect will be obtained. The width *w*, depth R and pitch *DL* of the recesses shown in Figure 3—1 and the width

w, height R and pitch DL of the ridges shown in Figure 4—1 depend upon the properties, especially melting point of a heat-and pressure-sensitive adhesive agent to be used for the sheet. In addition, the provision of recesses or ridges on the surface of the adhesive layer is achieved by providing in advance such ridges or recesses of predetermined width and height or depth to a release sheet of paper or synthetic resin generally used for protection of the adhesive layer surface, applying said release sheet to a sheet coated with a heat-and-pressure-sensitive adhesive agent with said ridges or recesses contacted with the surface of said adhesive layer and bonding them together under heat and pressure, or coating the ridged or recessed surface of the release sheet with a heat-and pressure-sensitive adhesive agent and applying it to the back of the sheet for transfer. In either case, the release sheet will subsequently be stripped from the adhesive layer.

Shown below are experimental examples in which air impermeable sheets each formed with an adhesive layer provided with ridges or recesses of particular cross-sectional shape, depth, width and pitch, were prepared by applying thereto a rubber type heat- and pressure-sensitive adhesive agent (melting point: 70°C—80°C) and such sheet was bonded to a melamine-resin coated iron plate under heat and pressure at 90°C within a vacuum applicator in the usual manner.

Example 1.

With a fixed ridge height R of 45μ, six different ridge widths w of 50μ, 90μ, 130μ, 260μ, 500μ, and 1000μ, and four different pitches DL of 2 cm, 4 cm, 6 cm and 8 cm for each of said widths, 24 release sheets having ridges of arcuate cross-section were prepared. Each of said release sheets was pressed under heat against a heat-and pressure-sensitive adhesive sheet coated with a heat-and pressure-sensitive adhesive agent having a dry film thickness of 50μ in such a manner that the ridged surface of said release sheet was contacted with the adhesive layer, thereby preparing 24 air impermeable heat-and pressure-sensitive sheets having arcuate recesses on the surface of said adhesive layer. These 24 experimental samples were bonded to objects under heat and pressure and the presence of visible air bubbles on the sheet receiving surfaces was observed and judged, the results of which are shown in Table 1.

Width (w)	Pitch (DL) 2 cm	4 cm	6 cm	8 cm
50	x	x	x	x
90	Δ	x	x	x
130	0	0	Δ	x
260	0	0	0	Δ
500	0	0	0	Δ
1000	0	0	0	Δ

(Notes) x . . . . . Large air bubbles

Δ. . . . . Trace amount of air bubbles

0 . . . . . Completely degassed

As seen in Table 1, it has been found that in the case where the height of the ridges of arcuate cross-section on the release sheet is 45μ, i.e., the depth of the recesses in the surface of the adhesive layer is 45μ, complete degassing is possible provided

that the pitch DL is 6 cm or below and the width w is greater than 130μ. When the width w was 1000μ, however, the recesses were visible from the surface of the sheet after the sheet was bonded, this being undesirable.

Example 2.

With a fixed pitch (DL) of 4 cm, five different recess depths R of 20μ, 40μ, 60μ, 80μ, and 100μ, four different widths (w) of 100μ, 200μ, 500μ and 1000μ for each of said depths, totalling 20 combinations, and three different cross-sectional shapes of recesses, arc, triangle and square, 60 release sheets having parallel recesses were prepared. The recessed surfaces of these release sheets were coated with the adhesive agent used in Experimental Example 1. The adhesive layers were applied to the backs of adhesive sheets for transfer to prepare 60 different air impermeable heat-and pressure-sensitive adhesive sheets having parallel ridges on their adhesive layers surfaces. They were bonded under heat and pressure to objects, the results of observation being shown in Table 2.

TABLE 2

Height (R)	Width (w)	Cross-sectional shape of ridges on adhesive layer		
		Arc	Triangle	Square
20μ	100μ	x	x	x
	200 ,,	x	x	x
	500 ,,	x	x	x
	1000 ,,	x	x	x
40μ	100μ	Δ	Δ	0
	200 ,,	Δ	x	0
	500 ,,	x	x	Δ
	1000 ,,	x	x	Δ
60μ	100μ	0	0	0
	200 ,,	0	Δ	0
	500 ,,	Δ	x	0
	1000 ,,	x	x	0
80μ	100μ	0	0	0
	200 ,,	0	0	0
	500 ,,	0	Δ	0
	1000 ,,	Δ	x	0
100μ	100μ	0	0	0
	200 ,,	0	0	0
	500 ,,	0	0	0
	1000 ,,	Δ	Δ	0

The results given in Table 2 show that in the case of sheets provided with parallel ridges spaced at intervals of 4 cm, there is substantially no difference in the results when the cross-section of the ridges is an arc whose height is 60μ or above and whose width is below 500μ, when it is a triangle whose height is 80μ or above and whose width is below 500μ; and when it is a square whose height is 40μ or above regardless of its width.

The air impermeable heat-and pressure-sensitive adhesive sheets of the present invention are constructed in the manner described above. They will not cause any trouble to the print on their surfaces. If they are provided on their adhesive layer surfaces with recesses having a depth, width and pitch or ridges having a height, width and pitch agreeing with the melting point of the heat-and pressure-sensitive adhesive agent used, there will be no possibility of air bubbles being trapped between the surface of the object and the adhesive layer surface when the sheets are bonded to the surfaces of objects under heat and pressure. Further, the surfaces of the sheets bonded are smooth and beautiful. In the case of reflecting sheets, there will be no loss of reflecting effect. Thus, the sheets are decidedly superior.

Embodiments of sheets of the present invention will now be described:

Embodiment 1:

A reflex-reflecting sheet having on the back thereof an air impermeable heat-and pressure-sensitive adhesive layer provided on its surface with arcuate recesses having a depth of 45μ, a width of 20μ and a pitch of 4 cm, the recesses intersecting an end edge of the sheet, was printed with a circle-contoured traffic sign designating "no parking". The sheet, which was circular, was bonded to a 60 cm diameter melamine resin coated plate under heat and pressure at 90°C using a vacuum applicator, thereby providing a traffic sign board. The printed surface of the sheet was more beautiful than that of a traffic sign board formed of a reflecting sheet having tiny holes separately prepared for comparison purposes, and the bonded surface was found to have been completely degassed, having no air bubbles noticeable.

Embodiment 2:

The surface of an air impermeable heat-and pressure-sensitive adhesive layer on the back of a reflex-reflecting sheet similar to the one used in Embodiment 1 was provided with parallel square ridges having a height of 60μ, a width of 150μ and a pitch of 4 cm, the ridges intersecting an end edge of the sheet, and a traffic sign board was obtained as in Embodiment 1. In this case also, the result was as satisfactory as in Embodiment 1.

WHAT WE CLAIM IS:—

1. An air impermeable heat-and pressure-sensitive adhesive sheet provided on its back surface with a heat-and pressure-sensitive adhesive layer, wherein the surface of said adhesive layer is provided with one or more elongate ridges or recesses and wherein at least one end of such ridge or recess intersects the end edge of the sheet.

2. A sheet as claimed in Claim 1, wherein said ridges or recesses are arranged in paralled and straight lines.

3. A sheet as claimed in Claim 1, wherein said ridges or recesses are arranged in parallel waves.

4. A sheet as claimed in Claim 1, wherein said ridges or recesses are arranged in a grid formation.

5. A sheet as claimed in any preceding Claim, wherein recesses of arcuate cross-section having a depth of 45μ and a width of 130μ—1000μ are arranged in parallel at intervals of up to 6 cm.

6. A sheet as claimed in any of Claims 1 to 4 wherein ridges of arcuate cross-section having a height of 60μ—100μ and a width of up to 500μ are arranged in parallel at intervals of 4 cm.

7. A sheet as claimed in any of claims 1 to 4, wherein ridges of triangular cross-section having a height of 80μ—100μ and a width of up to 500μ are arranged in parallel at intervals of 4 cm.

8. A sheet as claimed in any of Claims 1 to 4, wherein ridges of rectangular cross-section having a height of 40μ—100μ and a width of 100μ—1000μ are arranged in parallel at intervals of 4 cm.

9. A sheet as claimed in any preceding Claim, wherein said adhesive layer consists of a rubber type heat-and pressure-sensitive adhesive agent having a melting point of 70—80°C.

10. A method of producing a heat-and pressure-sensitive sheet as claimed in any preceding claim comprising providing a release sheet of paper or synthetic resin on one surface thereof with elongate ridges or recesses in advance, bonding under heat and pressure said release sheet to an air impermeable sheet provided on its back with a heat-and pressure-sensitive adhesive agent, in such a manner that the ridged or recessed surface of said release sheet is opposed to the adhesive layer, said release sheet being subsequently stripped from the adhesive layer.

11. A method of producing a heat-and pressure-sensitive adhesive sheet as claimed in any of Claims 1—9, comprising providing

a release sheet of paper or synthetic resin on one surface thereof with elongate ridges or recesses in advance, coating the ridged or recessed surface of said release sheet with a heat-and pressure-sensitive adhesive agent, applying said release sheet to the back of an air impermeable sheet with its adhesive layer opposed to the sheet to thereby transfer said adhesive layer to said sheet, said release sheet subsequently being stripped from the adhesive layer.

12. An air impermeable heat-and pressure-sensitive adhesive sheet substantially as herein described and illustrated.

13. A method of producing an air impermeable heat-and pressure-sensitive adhesive sheet substantially as herein described and illustrated.

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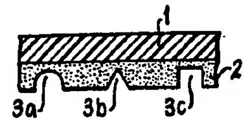

Fig. 1

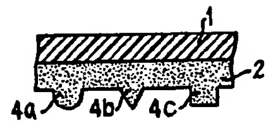

Fig. 2

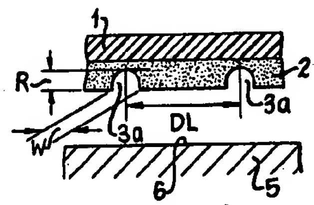

Fig. 3-1

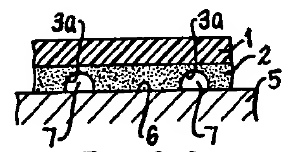

Fig. 3-2

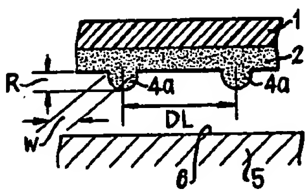

Fig. 4-1

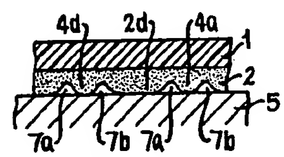

Fig. 4-2

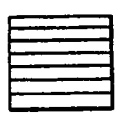

Fig 5-1

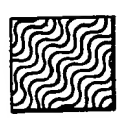

Fig. 5-2

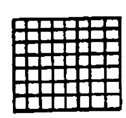

Fig. 5-3